

PATENT SPECIFICATION

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(54) THE REMOVAL OF SOLVENTS FROM SOLUTIONS

(71) We, SIEMENS AKTIENGESELLSCHAFT, a German Company, of Berlin and Munich, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to the removal of solvents from solutions, and is more particularly but not exclusively concerned with the separation of water of reaction from the electrolytes of fuel cells and fuel batteries.

Various methods of removing the water of reaction from the electrolyte of fuel cells and fuel batteries are possible. For example, a circulated reaction gas can be charged within the fuel cell with water evaporated from the electrolyte by the electrodes and this water can be removed outside the fuel cell in a condenser (see U.S. Patent Specification 3,462,308). A better water vapour saturation of the reaction gas can be achieved by effecting it outside the fuel cell, guiding the electrolyte into a concentrator in which the reaction water is absorbed in vapour form by a carrier gas (British Patent Specification No. 1,108,621).

Another method of removing the reaction water is described in the main Patent Specification No. 1,208,671. The electrolyte is brought into contact with a porous diaphragm, and water vapour diffusing therethrough is separated off on a cooled condensation surface; the hydrostatic pressure of the electrolyte in the diaphragm can be compensated for either by gas pressure or by capillary depression pressure. The separation of water of reaction in accordance with this diffusion-condensation principle has proved successful in practice. According to Austrian Patent Specification No. 277,341, the condensation surface may con-

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sist entirely or partially of a porous material. In this case, the water vapour which diffuses from the electrolyte through the porous diaphragm and across an adjacent gas chamber is condensed on the cooled condensation surface and forced by means of a compressed gas directly through the pores of the condensation surface and into the coolant. Alternatively, the cooled condensation surface, which bounds the said gas chamber, may be non-porous. In this case, the gas chamber, which may also be referred to as the condensation chamber, is provided with means for extracting the water of condensation. This means may comprise a pressure lock for the automatic continuous extraction of the water of reaction, into which lock the condensate is carried. Such a lock may comprise a porous plate through which the separated water of reaction is discharged (Patent Specification No. 1,111,316).

In the separation of water of reaction with the aid of a condensation chamber having a non-porous condensation surface and a sequentially disposed pressure lock, there may be employed for the transport of the condensate a pressurized auxiliary gas, for example hydrogen, which is extracted from the fuel cell or fuel battery. This auxiliary gas can be discharged into the ambient atmosphere after leaving the lock, which may involve a not inconsiderable consumption of gas. In addition, when hydrogen is employed as the auxiliary gas additional means may have to be provided to prevent the formation of explosive hydrogen-air mixtures.

According to one aspect of the present invention there is provided a method of removing solvent from a liquid solution, wherein the liquid solution is brought into contact with one face of a porous diaphragm such that, and under such conditions that, solvent vapour from the liquid

solution diffuses through the diaphragm and is condensed on a cooled non-porous condensation surface, the resultant effect of capillary and hydrostatic pressures of the liquid solution in the pores of the diaphragm being balanced with pressure of an auxiliary gas supplied to a condensation chamber that extends between and is bounded by the condensation surface and the other face of the diaphragm so that passage of the liquid solution through the porous diaphragm into the condensation chamber and passage of the auxiliary gas through the porous diaphragm into the liquid solution adjacent to the said one face are prevented, and wherein auxiliary gas and solvent condensed in the condensation chamber pass therefrom into a gas chamber of a pressure lock, which pressure lock also comprises a liquid chamber separated from the said gas chamber by a porous liquid-permeable membrane that is gas-tight when impregnated with the solvent, but through which solvent passes from the gas chamber to the liquid chamber, enabling such condensed solvent to be separated out of the said auxiliary gas which gas is, after such separation, conducted from the gas chamber of the lock to the suction intake of a jet pump, the auxiliary gas supplied to the said condensation chamber being taken from a duct portion connected in series with the main passageway through the jet pump so that difference in gas pressure between the said suction intake and the said duct portion, due to operation of the said jet pump by the passage of such gas therethrough along the said main passageway serves to bring about the passage of the auxiliary gas and the condensed solvent from the condensation chamber to the said gas chamber of the pressure lock.

According to another aspect of the present invention there is provided a device for effecting removal of solvent from a liquid solution, comprising a solution chamber adapted to have the liquid solution passed therethrough and bounded by one face of a porous diaphragm, and a condensation chamber which extends between, and is bounded by, the other face of the diaphragm and a non-porous condensation surface which is provided with means for cooling it, the device being such that when it is in use, with solvent vapour from such solution in the solution chamber diffusing through the diaphragm and being condensed on the cooled condensation surface, the resultant effect of capillary and hydrostatic pressures of the liquid solution in the pores of the diaphragm can be balanced with pressure of an auxiliary gas supplied to the said condensation chamber so that passage of the liquid solution

through the porous diaphragm into the condensation chamber and passage of the auxiliary gas through the porous diaphragm into the liquid solution adjacent to the said one face are prevented, and the device further comprising a pressure lock, having a gas chamber and having a liquid chamber separated from the said gas chamber by a porous liquid-permeable membrane that is gas-tight when impregnated with solvent, a passageway arranged for conducting condensed solvent, and auxiliary gas, from the condensation chamber to the said gas chamber of the pressure lock, thereby enabling the condensed solvent to be separated out of the auxiliary gas so that the condensed solvent, after such separation, can be extracted from the device separately from such gas when the device is in use, and a jet pump having connected to its suction intake a passageway arranged for conducting auxiliary gas to that intake from the gas chamber of the pressure lock, the main passageway through the jet pump being in series with a duct portion from which a connection is provided to the said condensation chamber so that when the device is in use, with the jet pump being operated by having such gas passed therethrough along its main passageway, a pressure difference established between the said duct portion and the said suction intake by virtue of the operation of the jet pump serves to bring about passage of such condensed solvent, and auxiliary gas supplied to the condensation chamber by way of the said connection, to the said gas chamber of the pressure lock.

For example, in one application of the invention to the removal of water of reaction from the electrolyte of a fuel cell or battery, the jet pump is connected in a reaction gas supply line of the fuel cell or fuel battery so as to establish such a pressure difference, between the condensation chamber and the pressure lock, for transporting the condensate, and auxiliary gas transported together with the condensate is returned into the said supply line by the jet pump. When the fuel cell or battery is in operation, reaction gas (fuel and/or oxidising agent) is consumed. The gas flow along the main passageway through the jet pump produces a pressure drop at a nozzle region thereof where the suction intake of the jet pump is located. The difference between the pressure of the gas before it enters the nozzle region of the jet pump and the generated low pressure after it leaves the nozzle is utilised to transport the condensate out of the condensation chamber and into the lock. Auxiliary gas flowing with the condensate returns into the suction intake of the jet pump.

There may with advantage be employed as the auxiliary gas a reaction gas of the fuel cell battery, because in this case no additional gas need be present. If the fuel cell or the fuel battery is operated with hydrogen as fuel, the hydrogen is preferably used as the auxiliary gas.

Preferably the condensation chamber is connected to the appropriate gas supply line of the fuel cell or battery, while the lock is connected to the jet pump. In this case, the jet pump pressure differential alone serves for conveying the condensate out of the condensation chamber and into the pressure lock. In a further form of construction, the condensation chamber may be connected to a duct for the discharge of the gas from the fuel cell or fuel battery, and the lock may be connected to the jet pump. In this case, the jet pump and the gas serve both to flush the fuel cell or battery and — after the gas has left the fuel cell or battery — to flush the condensation chamber and the lock.

Reference will now be made, by way of example, to the accompanying diagrammatic drawings, in which Figures 1, 2, and 3, are schematic illustrations of respective first, second, and third, embodiments of the invention.

In Figure 1, a battery consisting of fuel cells is denoted by 1; the individual fuel cells of the battery are not illustrated in the drawing. The fuel battery 1 is supplied, through a supply line 2, with pressurised hydrogen as fuel; the supply source and the gas ducts in the battery are not shown. The electrolyte is pumped out of a supply vessel 3 with the aid of an electrolyte pump 4 through the fuel battery 1, in which it flows in parallel through the individual fuel cells in the manner indicated, and into a water removal unit 5, in which both the temperature and the concentration of the electrolyte may be regulated. The water removal unit 5 contains two electrolyte chambers 6, which are traversed in parallel by the electrolyte liquid. The electrolyte chambers 6 are bounded entirely or partially by porous diaphragms 7, for example in the form of asbestos membranes. The electrolyte chambers 6 are adjoined by condensation chambers 8, which are bounded by non-porous condensation surfaces 9, for example nickel sheets or plastics foils. The condensation surfaces 9 form at the same time partitions from adjacent cooling chambers 10, through which a coolant, for example water, flows by way of ducts 26 and 27 in the indicated manner.

The water removal unit may alternatively be of simpler construction and may comprise, for example, only one electrolyte chamber, one condensation chamber

and one cooling chamber.

The electrolyte and cooling chambers may be lined with gauzes for example of polypropylene. Temperature regulation of the electrolyte in the water removal unit may be effected by appropriately controlled cooling by means of a cooling liquid such as water. For regulating the concentration of the electrolyte liquid, there is employed a concentration-measuring device 11 which acts on a three-way valve 12. By means of this valve, water removed from a pressure lock 13 is either returned into the supply vessel 3 through a duct 25 or discharged through a duct 24, depending upon the concentration of the electrolyte. The concentration-measuring device 11 adjoins a duct 20, leading back to the supply vessel 3, of the electrolyte circuit.

Disposed in the gas supply line 2 of the fuel battery 1 is the main passageway of a jet pump 14. Hydrogen is taken off from the line 2 at a point short of the jet pump 14 and supplied to the water removal unit 5, in which it flows in parallel through the condensation chambers 8. Water vapour which diffuses through the diaphragms 7 from the electrolyte flowing through the electrolyte chambers 6 is precipitated upon the non-porous condensation surfaces 9 and the condensate collects in the lower part of the condensation chambers 8. For the transport of the condensate into the lock 13 together with the hydrogen present as auxiliary gas in the condensation chambers 8 by virtue of the pressure differential maintained by virtue of the operation of the jet pump 14, there is employed a collecting duct 16, which leads into a gas chamber 17 of the lock 13. The gas chamber 17 is separated from a liquid chamber 19 by a porous membrane 18, for example an asbestos membrane. The liquid chamber may be lined by a gauze, for example of polypropylene. The condensate collecting in the gas chamber 17 is forced through the diaphragm 18 into the liquid chamber 19, which is substantially at atmospheric pressure, and then either discharged with the aid of a valve 12 or returned into the electrolyte supply vessel 3 through a duct 25. Finally, the auxiliary gas returns into the suction intake 22 of the jet pump 14 from the gas chamber 17 through a duct 21, in which there is provided a non-return valve 23.

The apparatus operates as follows: when the battery is set in operation so that it is electrically loaded, but the cooling is not yet started in the water removal unit, a flow of pure hydrogen takes place through the duct 15, the condensation chambers 8 of the water removal unit 5, the duct 16, the gas chamber 17 of the lock 13, the

duct 21 and the non-return valve 23 to the jet pump 14. The rate of flow depends upon the flow resistances.

When the water removal unit is set in operation by starting of the cooling, a flow of gas liquid into the lock occurs with small quantities of condensate in the condensation chambers 8, and a corresponding liquid level is set up in the said lock. The rate of flow of the hydrogen is in this case determined substantially by the two-phase transport in the duct 16 to the lock 13 and the maintenance of the liquid level and it is relatively low in relation to the pure gas flow. As the quantity of condensate increases, there is obtained in the limit case a pure liquid flow from the condensation chambers into the lock. The pressure differential due to operation of the jet pump then serves only to maintain the liquid level in the lock. In this case, the highest liquid level is obtained in the lock and at the same time the maximum possible current of transported condensate.

When the battery is switched off, a liquid level will generally be present in the lock 13. If the pressure differential produced by operation of the jet pump 14 were then removed by interruption of the supply of fuel, an equalisation of the liquid level between the condensation chambers and the lock might occur, and possibly also contact of the condensate with the diaphragms of the electrolyte chambers in the water removal unit. Part of the condensate would then be forced back into the electrolyte circuit. This is prevented by the non-return valve 23, which maintains the relatively low pressure in the chamber 17.

If the duct 15 for the auxiliary gas branches off from the gas supply line 2 at a point short of the jet pump 14 as in Figure 1, the auxiliary gas passes through the jet pump only into the battery after flowing through the water removal unit and the lock. On the other hand, if the duct 15 is connected at a point downstream of the jet pump, the auxiliary gas, after flowing through the water removal unit and the lock, can pass through the jet pump both into the battery and again into the water removal unit and the lock.

Figure 2 diagrammatically illustrates a particularly preferred modified form of an apparatus similar to that of Figure 1. In this case, the lock and the water removal unit are flanged to the fuel battery, i.e. directly connected thereto. In the first place, this has the advantage that a compact constructional unit can thereby be obtained, in which ducts which must be provided between the individual parts when they are separately arranged are not required. Moreover, with such a compact constructional assembly some of the end

plates otherwise needed may be omitted, whereby the construction is simplified and a reduction in weight is obtained. Finally, with this compact construction, it is possible in the case of high output fuel batteries to effect a simple parallel connection of a number of pressure locks. In this case it is advantageous that the liquid level in the lock or locks is situated above the lower ends of the diaphragms in the water removal unit. With a separate arrangement of the individual parts, similar advantage can be obtained by disposing the lock at a lower level than the water removal unit.

The electrolyte supply vessel together with the associated ducts, and the electrolyte pump, have been omitted from Figure 2. The electrolyte liquid, for example 6 n KOH, passes through a duct 32 at a lower edge of the end plate 60 into the fuel battery 31, flows in parallel through all the fuel elements of the battery (not shown), as indicated in the Figure, and leaves an upper edge of the fuel battery 31 by way of a duct 33 in a separating plate 34 between the battery 31 and the flanged-on water removal unit 35, and enters a reversing cell 36 in the water removal unit 35. The direction of the electrolyte is reversed by means of the said cell 36, and it enters the duct 59 and then flows — from the bottom upwards — in a parallel direction through the electrolyte chambers 37 of the water removal unit 35. Thereafter, the electrolyte passes by way of a duct 38 out of the end plate 39 of the compact constructional unit consisting of the fuel battery, the water removal unit and the lock, and is finally returned into the electrolyte supply vessel. As is illustrated in Figure 1, a concentration-measuring device may be provided in the electrolyte circuit. A removal of water also takes place at the reversing cell 36.

A pressurised gaseous reactant, for example hydrogen under a pressure of 0.15 N/mm², is supplied through a duct 40 from a supply container (not shown) to the fuel battery 31. A jet pump 41 is disposed in the said supply duct 40. The jet pump may consist, for example, of glass. The internal diameter at its main inlet and outlet is, for example, about 6 mm, while the nozzle and the succeeding constriction have an internal diameter of, for example, about 0.7 mm. The reactant which is not reacted in the electrochemical reaction in the fuel battery 31 leaves the battery by way of a duct 42. Gaseous reactant, as auxiliary gas, is removed from the supply duct 40 for the reactant by way of a duct 43 at a point short of the jet pump 41 and fed to the condensation chambers 44 of the water removal unit 35. The auxiliary gas passes through the collecting duct 48 into two gas

chambers 49 of the lock 50 together with the condensate in the condensation chambers 44 which has been formed by diffusion of the water vapour from the electrolyte liquid in the electrolyte chambers 37 and the reversing cell 36 through the diaphragm 45 and condensation on the nonporous condensation surfaces 46 of the cooling chambers 47. The auxiliary gas leaves the top of the gas chambers 49 and is fed, through a common duct 51 in which a non-return valve 52 is provided, to the suction intake 53 of the jet pump 41. The condensate transported into the gas chambers 49 of the lock 50 by the pressure differential produced by operation of the jet pump 41 passes through the membranes 54 which bound the gas chambers 49 into a liquid chamber 55 and is removed from the latter through a duct 56. This advantageously takes place in the upper part of the battery, because drying of the membranes is thereby prevented. The condensate, i.e. the water, may either be discharged through the said duct 56 and a three-way valve as illustrated in Figure 1, or returned into the electrolyte supply container. The cooling medium for the water removal unit, for example water, passes from the bottom through the end plate 39 by way of a duct 57 into the water removal unit 35, flows in parallel from the bottom upwards through the cooling chambers 47 and leaves the compact constructional unit through a collecting duct 58 in the end plate 39.

There is diagrammatically illustrated in simplified form in Figure 3 a form a construction of an apparatus, in which on the one hand a water removal unit, having a condensation chamber 64, and a pressure lock 65 are combined in a compact constructional unit, and in which the condensation chamber is connected to receive its auxiliary gas input from the gas discharge duct 66 of a fuel battery 63. Most of the electrolyte circuit of the battery, the electrolyte supply vessel and the electrolyte pump, and the supply vessel for the gaseous reactant have been omitted for the sake of simplicity. The gaseous reactant flows through a duct 61 and a jet pump 62 into the fuel battery 63, in which it flows in suitable manner, for example in parallel or in series, through the fuel cells (not shown), and from there through the duct 66 into the condensation chamber 64 and through a duct 67 into a gas chamber 68 of the lock 65. The reactant serving as auxiliary gas is therefore derived directly from the gas outlet of the battery, which may be operated with increased gas throughput. The gas outlet of the lock 65 is directly connected to the suction intake of the jet pump 62 by a duct 69. The elec-

trolyte liquid passes through a duct 70 into the battery, flows through the individual fuel cells, which are not shown, and through a duct 71 into an electrolyte chamber 72 which is separated from the condensation chamber 64 by a diaphragm 73, and leaves the electrolyte chamber 72 by way of a duct 74. A coolant passes through a duct 75 into a cooling chamber 76, which is separated from the condensation chamber 64 by a non-porous condensation surface 77, and leaves the cooling chamber by way of a duct 78. A liquid chamber 79 of the lock 65 is separated from a gas chamber 68 thereof by a liquid-permeable membrane 80. Water collecting in the liquid chamber 79 is removed from the lock through a duct 81.

Experiments have shown that even with battery outputs of the order of magnitude of 100 watts, the pressure difference required for the transport of the condensate can be obtained. In the case of higher battery outputs, simple jet pumps can be sufficient, especially if there are employed, in the pressure locks, membranes having good permeability (for example leather-asbestos membranes of a thickness of about 0.4 mm). Since, in accordance with Figures 1 to 3, the auxiliary gas used for the transport of the condensate is returned into the gas supply duct of the battery on leaving the lock, no gas losses need occur in the delivery of the condensate, so that the efficiency of the whole battery can be increased.

WHAT WE CLAIM IS:—

1. A method of removing solvent from a liquid solution, wherein the liquid solution is brought into contact with one face of a porous diaphragm such that, and under such conditions that, solvent vapour from the liquid solution diffuses through the diaphragm and is condensed on a cooled non-porous condensation surface, the resultant effect of capillary and hydrostatic pressures of the liquid solution in the pores of the diaphragm being balanced with pressure of an auxiliary gas supplied to a condensation chamber that extends between and is bounded by the condensation surface and the other face of the diaphragm so that passage of the liquid solution through the porous diaphragm into the condensation chamber and passage of the auxiliary gas through the porous diaphragm into the liquid solution adjacent to the said one face are prevented, and wherein auxiliary gas and solvent condensed in the condensation chamber pass therefrom into a gas chamber of a pressure lock, which pressure lock also comprises a liquid chamber separated from the said gas chamber by a porous liquid-permeable membrane that is gas-tight when im-

pregnated with the solvent but through which solvent passes from the gas chamber to the liquid chamber, enabling such condensed solvent to be separated out of the said auxiliary gas which gas is, after such separation, conducted from the gas chamber of the lock to the suction intake of a jet pump, the auxiliary gas supplied to the said condensation chamber being taken from a duct portion connected in series with the main passageway through the jet pump so that difference in gas pressure between the said suction intake and the said duct portion, due to operation of the said jet pump by the passage of such gas therethrough along the said main passageway, serves to bring about the passage of the auxiliary gas and the condensed solvent from the condensation chamber to the said gas chamber of the pressure lock.

2. A device for effecting removal of solvent from a liquid solution, comprising a solution chamber adapted to have the liquid solution passed therethrough and bounded by one face of a porous diaphragm, and a condensation chamber which extends between, and is bounded by, the other face of the diaphragm and a non-porous condensation surface which is provided with means for cooling it, the device being such that when it is in use, with solvent vapour from such solution in the solution chamber diffusing through the diaphragm and being condensed on the cooled condensation surface, the resultant effect of capillary and hydrostatic pressures of the liquid solution in the pores of the diaphragm can be balanced with pressure of an auxiliary gas supplied to the said condensation chamber so that passage of the liquid solution through the porous diaphragm into the condensation chamber and passage of the auxiliary gas through the porous diaphragm into the liquid solution adjacent to the said one face are prevented, and the device further comprising a pressure lock, having a gas chamber and having a liquid chamber separated from the said gas chamber by a porous liquid-permeable membrane that is gas-tight when impregnated with solvent, a passageway arranged for conducting condensed solvent, and auxiliary gas, from the condensation chamber to the said gas chamber of the pressure lock, thereby enabling the condensed solvent to be separated out of the auxiliary gas so that the condensed solvent, after such separation can be extracted from the device separately from such gas when the device is in use, and a jet pump having connected to its suction intake a passageway arranged for conducting auxiliary gas to that intake from the gas chamber of the pressure lock, the main passageway through the jet pump being in

series with a duct portion from which a connection is provided to the said condensation chamber so that when the device is in use, with the jet pump being operated by having such gas passed therethrough along its main passageway, a pressure difference established between the said duct portion and the said suction intake by virtue of the operation of the jet pump serves to bring about passage of such condensed solvent, and auxiliary gas supplied to the condensation chamber by way of the said connection, to the said gas chamber of the pressure lock.

3. A device as claimed in claim 2, wherein the said porous liquid-permeable membrane is an asbestos membrane.

4. A device as claimed in claim 2 or 3, connected with a fuel cell so as to remove water of reaction (being the said solvent) from the fuel cell electrolyte (being the said liquid solution) when the device and the fuel cell are in use.

5. A device as claimed in claim 4, connected to employ as the said auxiliary gas a reaction gas of the fuel cell.

6. A device as claimed in claim 5, wherein the said reaction gas is hydrogen.

7. A device as claimed in claim 5 or 6, wherein the said main passageway forms part of a reaction gas supply line of the fuel cell so that the said reaction gas is delivered to a reaction gas inlet of the fuel cell from the outlet of the jet pump when the cell is in operation.

8. A device as claimed in claim 5, 6, or 7, wherein the condensation chamber is connected to receive the said auxiliary gas, from part of a reaction gas supply line leading to a reaction gas inlet of the fuel cell, before that auxiliary gas is passed through the fuel cell when in operation.

9. A device as claimed in claim 5, 6, or 7, wherein the condensation chamber is connected to receive the said auxiliary gas, from an outlet of the fuel cell, after that gas has passed through the fuel cell when in operation.

10. A device as claimed in any one of claims 4 to 9, wherein the said fuel cell is part of a battery comprising a plurality of such cells, having respective electrolyte chambers connected together in an electrolyte circuit including the said solution chamber.

11. A device as claimed in any one of claims 2 to 10, wherein the pressure lock and the condensation chamber are provided in respective structural units which are flanged together.

12. A device as claimed in claim 11 read as appendant to any one of claims 4 to 10, wherein the said structural units are flanged together with a housing of the fuel cell

13. A device, including a jet pump, for effecting removal of solvent from a liquid solution, substantially as hereinbefore described with reference to Figure 1, 2, or 3, 5 of the accompanying drawings.

14. A combination of a fuel cell battery and a device, including a jet pump, for removing water of reaction from the electrolyte of the battery, substantially as hereinbefore described with reference to Figure 10 1, 2, or 3, of the accompanying drawings.

15. A method of removing solvent from a liquid solution, employing a jet pump in

a manner substantially as hereinbefore described with reference to Figure 1, 2, or 3, 15 of the accompanying drawings.

16. A method as claimed in claim 1, wherein there is employed a device as claimed in any one of claims 3 to 13.

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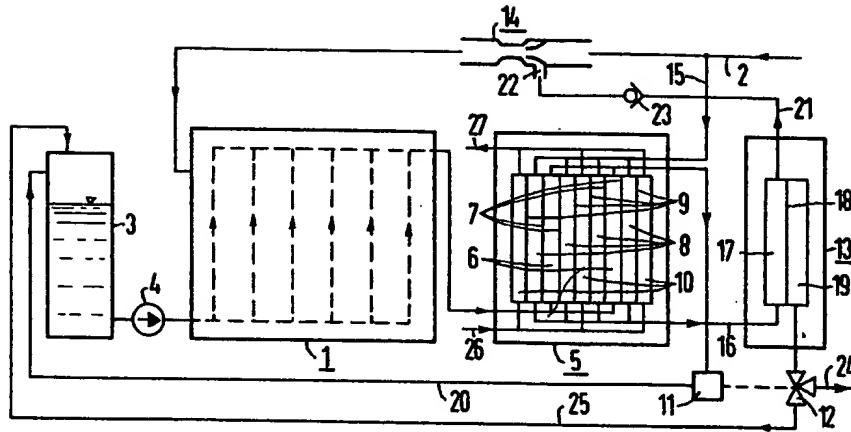


Fig. 1

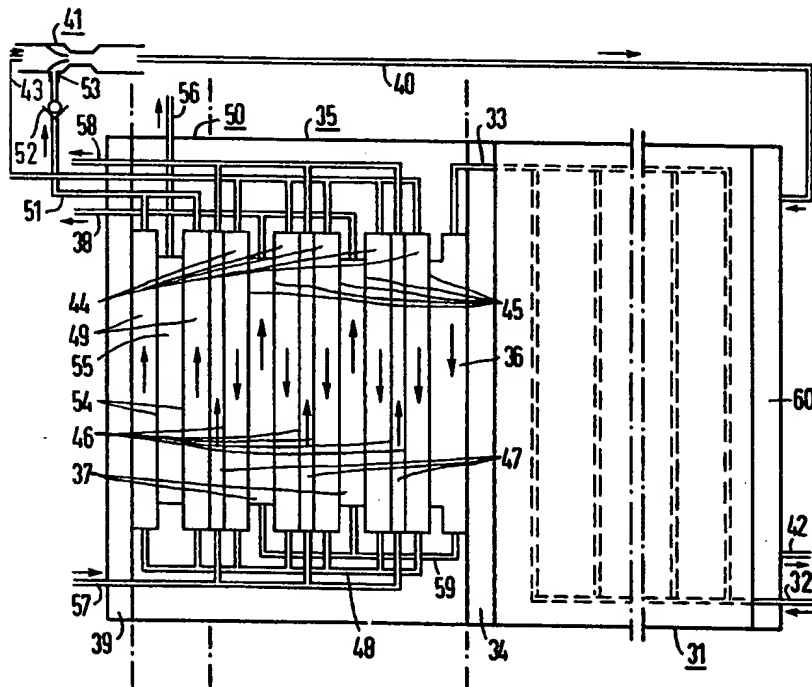


Fig. 2

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2 SHEETS

2 SHEETS

COMPLETE SPECIFICATION

**This drawing is a reproduction of
the Original on a reduced scale.
SHEET 2**

SHEET 2

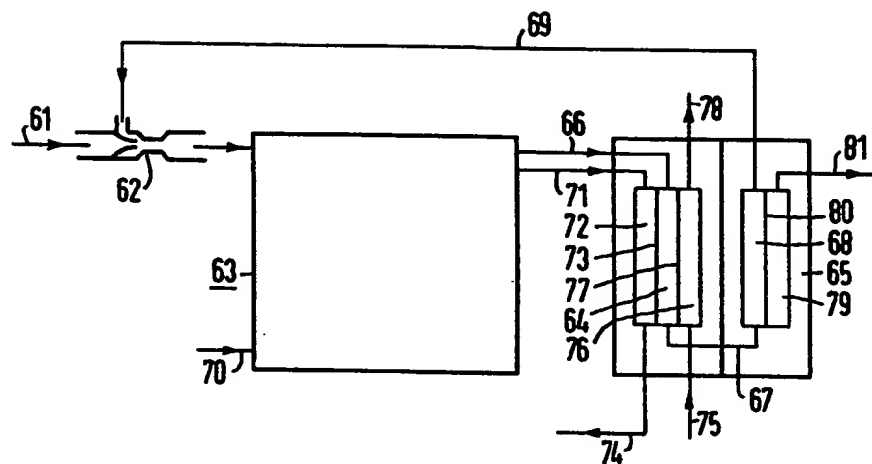


Fig.3